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(54) A method for the preparation of a cellulose ether having a decreased degree of polymerization.

(57) The invention provides a method for preparing a water-soluble cellulose ether of high whiteness having a decreased average degree of polymerization suitable as a film-coating agent of solid medicament forms free from the problem of noxious impurities. The method comprises contacting the starting cellulose ether with an aqueous solution of hydrogen chloride in an amount limited in respect to the amounts of hydrogen chloride and water at 40 to 85 °C for a length of time to effect molecular weight decrease followed by dissipation of hydrogen chloride and water.

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A METHOD FOR THE PREPARATION OF A CELLULOSE ETHER HAVING A DECREASED DEGREE OF POLYMERIZATION

BACKGROUND OF THE INVENTION

The present invention relates to a method for the preparation of a cellulose ether having a decreased average degree of polymerization from a starting cellulose ether having a higher degree of polymerization. More particularly, the invention relates to a method for the preparation of a water-soluble cellulose ether of high whiteness having such a decreased average degree of polymerization that a 2 % by weight aqueous solution thereof has a viscosity of 20 centipoise or lower at 20 °C starting from a cellulose ether having a higher degree of polymerization.

As is known, water-soluble cellulose ethers having a relatively low average degree of polymerization or molecular weight are useful, for example, as a film-coating material on solid medicament forms, e.g. pills and tablets, binder of such solid medicament forms, thickening agent of various pasty materials and so on. Several methods are proposed and industrially undertaken for the preparation of such low-molecular water-soluble cellulose ether products. For example, the etherification reaction of an alkali cellulose, which is obtained by the reaction of sodium hydroxide on the starting cellulose pulp, is preceded by a process of air oxidation to give a low-molecular alkali cellulose to be

whiteness without the above described problems and disadvantages in the prior art methods. The object of the invention is, particularly, to provide a method for the preparation of a cellulose ether product which gives a 2 % by weight aqueous solution having a viscosity of 20 centipoise or lower at 20 °C.

Thus, the method of the present invention for the preparation of a water-soluble cellulose ether having a decreased average degree of polymerization comprises:
(a) contacting a starting cellulose ether in a powdery form with an aqueous solution of hydrogen chloride in such an amount that the amount of the hydrogen chloride is in the range from 0.1 to 1.0 % by weight based on the starting cellulose ether and the amount of water is in the range from 3 to 8 % by weight based on the overall amount of the starting cellulose ether and the aqueous solution of hydrogen chloride at a temperature in the range from 40 to 85 °C; and
(b) removing the hydrogen chloride from the mixture of the powdery cellulose ether and the aqueous solution of hydrogen chloride.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

The above described method of the present invention is applicable to a variety of water-soluble cellulose ethers as the starting material including alkyl and hydroxyalkyl celluloses such as methyl cellulose, hydroxyethylcellulose,

achieved only by taking an unduly long time. When the amount of hydrogen chloride is too large, on the other hand, the reaction velocity would be so large that good control of the reaction can hardly be achieved in addition to the problem that the resultant product can be freed from residual hydrogen chloride only by a long time of removing procedure or may contain an increased amount of chloride ions as an impurity even after a prolonged procedure of removal.

Another important factor in the treatment is the amount of water contained in the mixture of the starting cellulose ether and the aqueous solution of hydrogen chloride. Namely, the water content in the mixture should be in the range from 3 to 8 % by weight or, preferably, from 5 to 8 % by weight based on the overall amount of the starting cellulose ether and the aqueous solution of hydrogen chloride. When the amount of water is too small corresponding to a high concentration of hydrogen chloride in the solution, the reaction velocity is somewhat increased accordingly but the resultant product is sometimes colored in yellow. When the amount of water is too large, on the other hand, agglomerates of the cellulsce ether particles may be formed in addition to the decrease in the reaction velocity taking an unduly long time for completion of the reaction. A commercially available product of cellulose ethers usually contains from about 0.5 to about 2.5 % by weight of moisture so that this amount of water in the starting cellulose ether should be taken into

reaction is permissible in the product.

According to the inventive method, the degree of polymerization can be efficiently decreased in a simple and convenient process without the disadvantages of noticeable yellowing and remaining impurities such as oxidizing agents, organic solvents, sulfur compounds and the like so that the cellulose ether products obtained by the inventive method are very useful as a coating agent of solid medicament forms of which high whiteness is particularly desirable.

In the following, the method of the present invention is described in more detail by way of examples, in which the viscosity of the aqueous solution of cellulose ethers is given with the value obtained by the measurement at 20 °C for a 2 % by weight aqueous solution in each occurrence.

Example 1 (Experiments No. 1 to No. 5).

In each of Experiments No. 2 to No. 5, into a Henschel mixer of 10 liter capacity was taken 1 kg of a powdery hydroxypropyl methyl cellulose, of which the aqueous solution had a viscosity of 210 centipoise, containing 10 % by weight of hydroxypropoxyl groups, 29 % by weight of methoxyl groups and 0.9 % by weight of moisture and the mixer was driven at a velocity of 200 rpm. An aqueous solution of hydrogen chloride in a varied concentration as indicated in Table 1 below was sprayed to the cellulose ether powder in the running

Table 1

Experiment No.	HCl concentration of acid solution, %	Overall moisture content, %	Reaction time, hours	Viscosity of aqueous solution, centipoise	Yellowness, YI
1	(dry gas)	0.9	1.6	6.1	21
2	20	2.1	2.0	5.7	17
3	10	3.5	2.0	6.4	13
4	5	6.2	3.0	6.7	13
5	3	9.6	5.0	7.7	13

50 mm optical path using a color computer (Model SM-2, manufactured by Suga Test Instruments Co.) to give the results shown in Table 1, in which Experiments No. 1, No. 2 and No. 5 were for comparative purpose and Experiments No. 3 and No. 4 were for the invention. In Experiment No. 5, some agglomerates were found in the cellulose ether powder taken out of the tumbling vessel.

Example 2.

The experimental procedure was substantially the same as in Example 1, in which 40 g of a 10 % aqueous solution of hydrogen chloride were sprayed to 1 kg of a powdery methyl cellulose, of which the aqueous solution had a viscosity of 380 centipoise, containing 29.7 % by weight of methoxyl groups and 1.2 % by weight of moisture and the powder was

For comparison, 500 g of the same ethyl cellulose powder were taken in a vessel which was tumbled in the same manner as above while 3.5 g of dry hydrogen chloride gas were blown thereinto over the length of time. The thus obtained powdery ethyl cellulose gave an aqueous solution having a viscosity of 3.5 centipoise but it was colored in clearly deeper yellow than above with a yellowness YI of 27.